Electronic Supplementary Material (ESI) for ChemComm.

Supporting Information

Observing the Phase Transformation of CVD-grown MoS_2 via Atomic Resolution TEM

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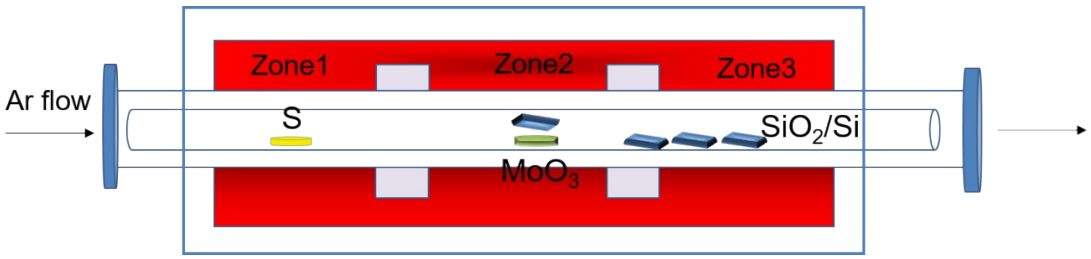


Figure S1. Schematic illustration of the CVD system setting.

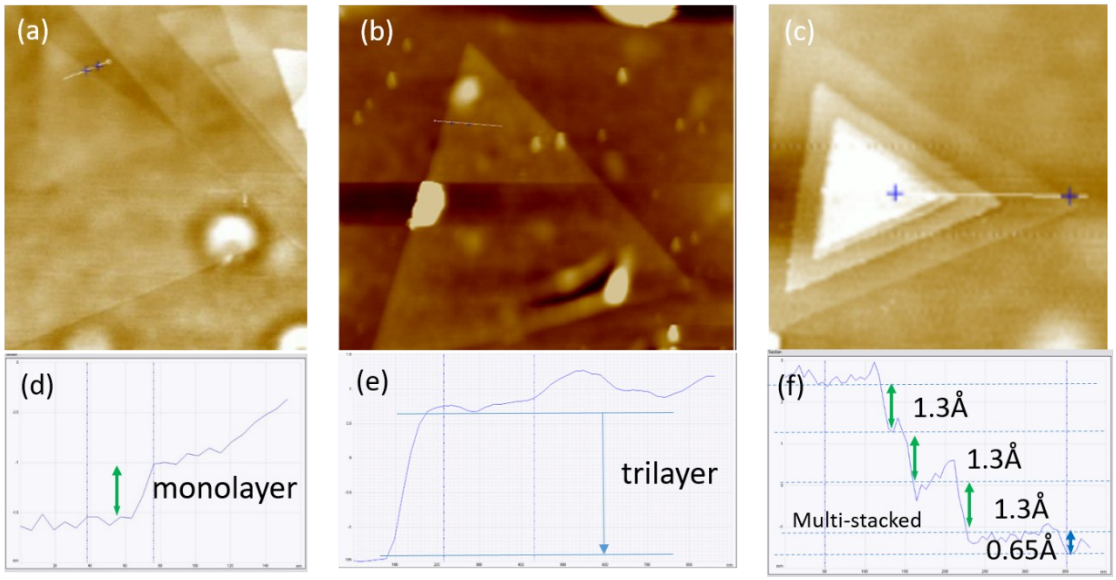


Figure S2. AFM image and height profile (a,d) AFM image shows the stacking layered MoS_2 and the height variation is 0.65nm (monolayer). (b,e) Another trilayer-growth MoS_2 with triangular domain and the height variation is 2.1nm(trilayer). (c) Multi-stacked pyramid MoS_2 was depicted in the AFM image. (f) Step height profile along the line indicated in (c).

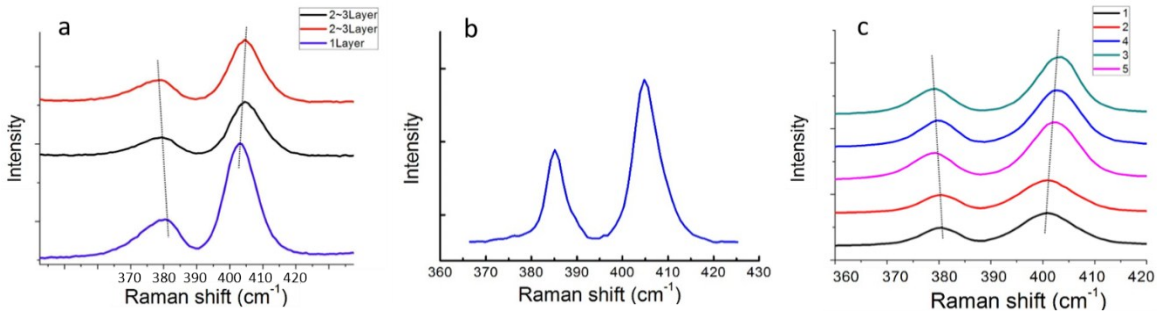


Figure S3. Raman spectroscopy measurement. (a) Raman shift correspond to the Figure 1(a)(b), which shows Δ=20cm^{-1}~monolayer. (b) Raman shift correspond to the Figure 1(d), showing the layer composed of monolayer to trilayer. (c) Raman-shift of the netted-film layered, which shows a multilayer result.

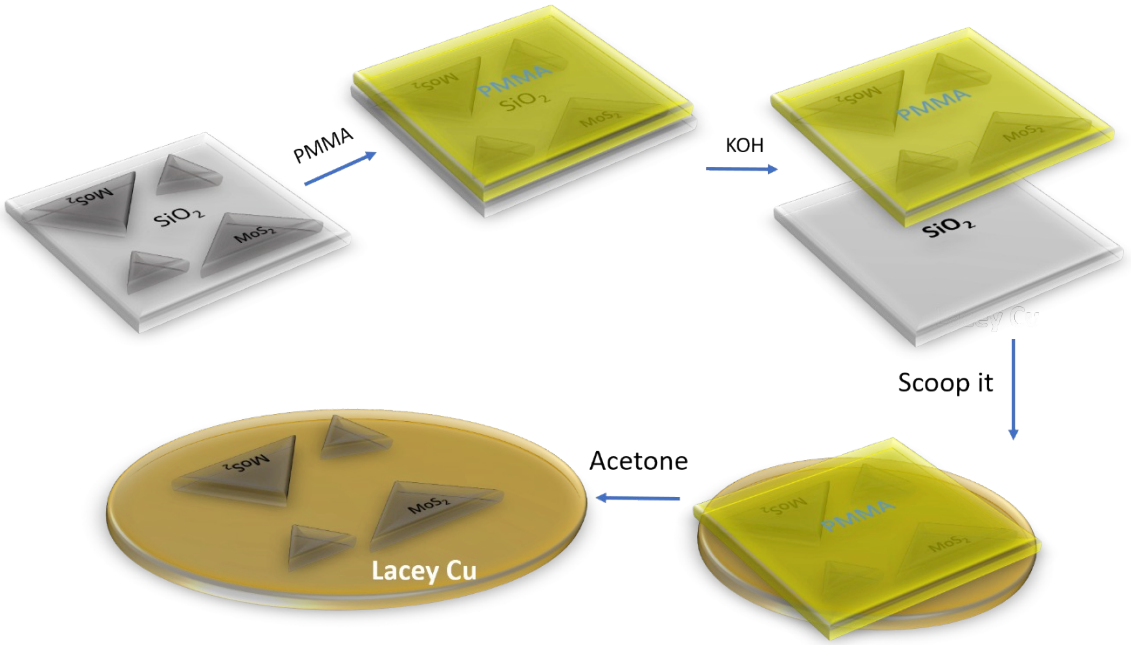


Figure S4. Illustration of transferring method.

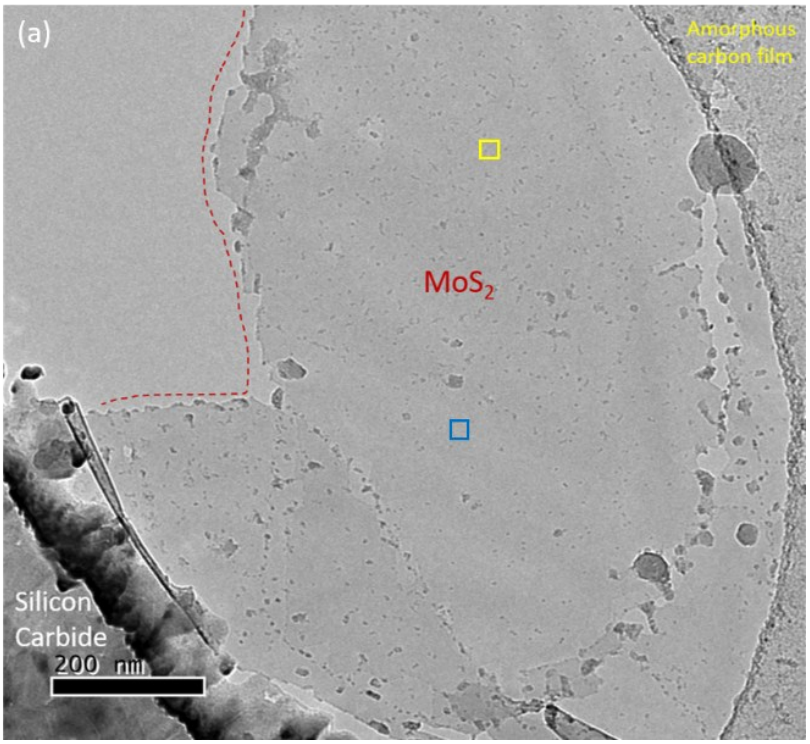
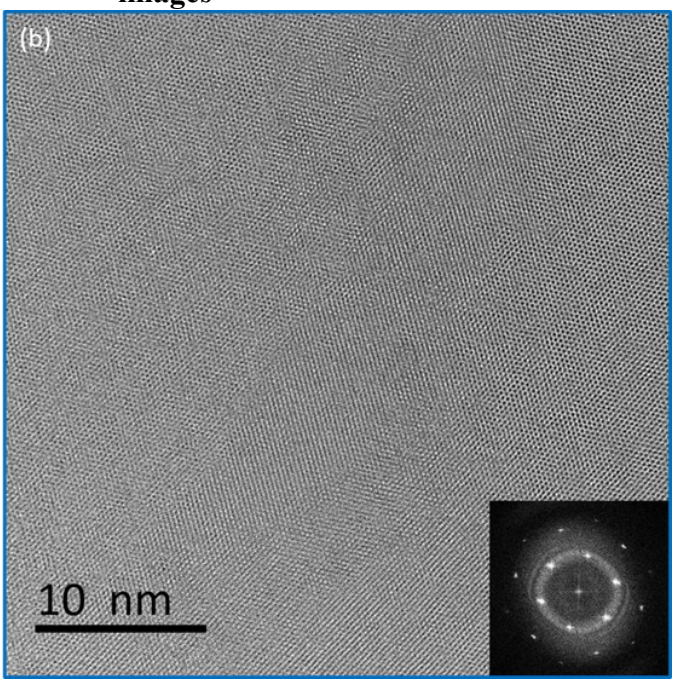
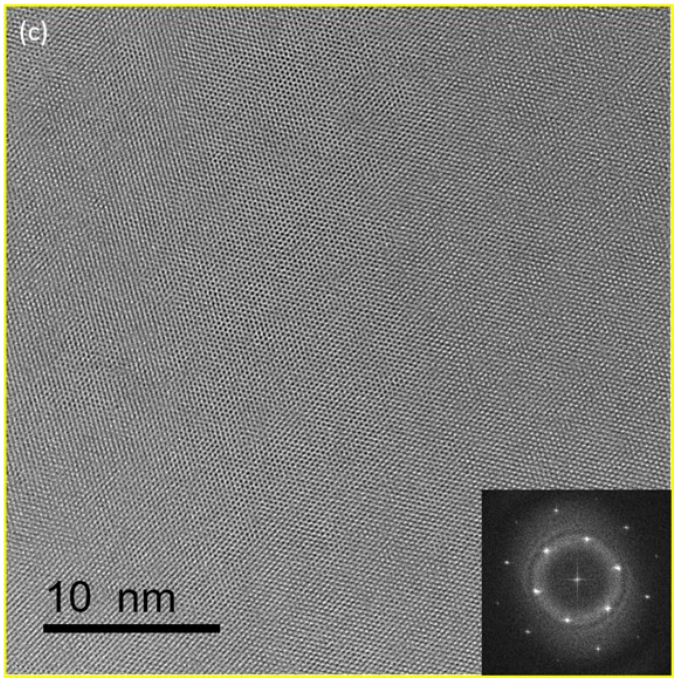


Figure S5. TEM images of the transferred MoS_2 layers.





(a) Large-area TEM image of transferred single-layer MoS_2 (some of the region overlapped). (b) HRTEM image of the as-grown sample highlighted with blue box in figure (a). (c) HRTEM image of the as-grown sample highlighted with yellow box in figure (a)

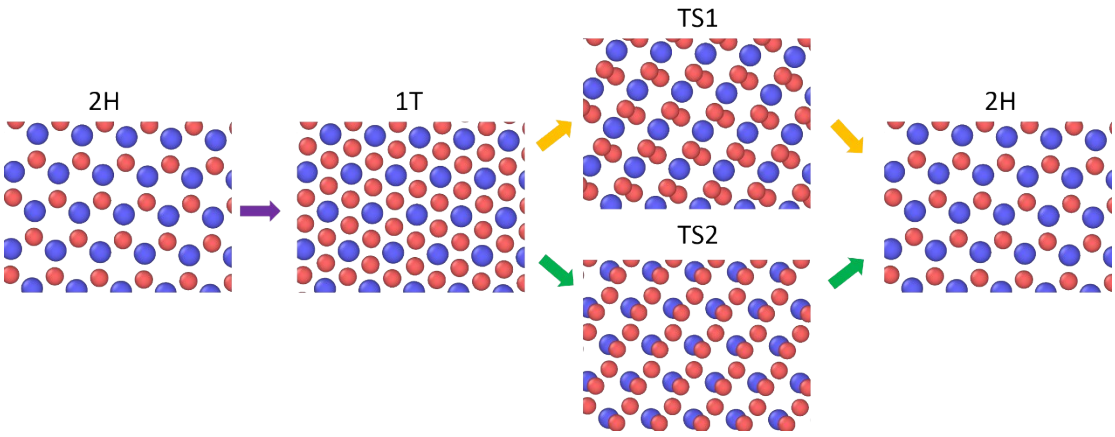


Figure S6. Illustrated model of the phase transition path.

We built up the atomic model for 2H, 1T, TS1 and TS2 phases, respectively. The red atoms represented sulfur atoms while the blue ones represented the molybdenum atoms. This transitional path were 2H-1T-TS1/TS2-2H and Movie S3/Movie S4 provide the dynamic information as reference.

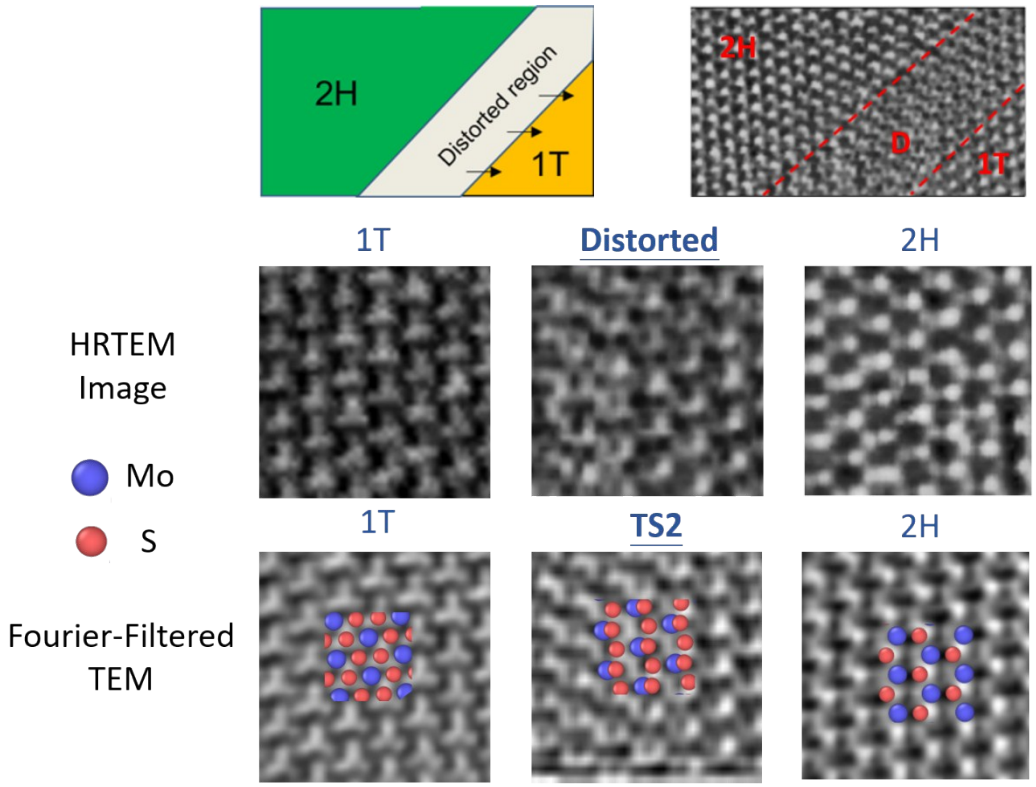


Figure S7. The comparison between the established model and the experimental evidence. The experimental image of 1T phase, the distorted region dragged out from figure 4c, and 2H phase. The Fourier-Filtered image were compared with the atomic model from figure S6.

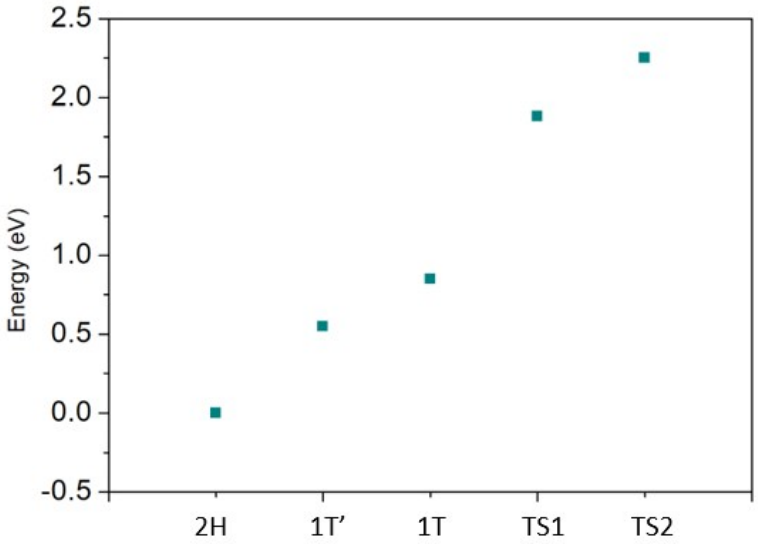


Figure S8. The plot of energy vs different phases.

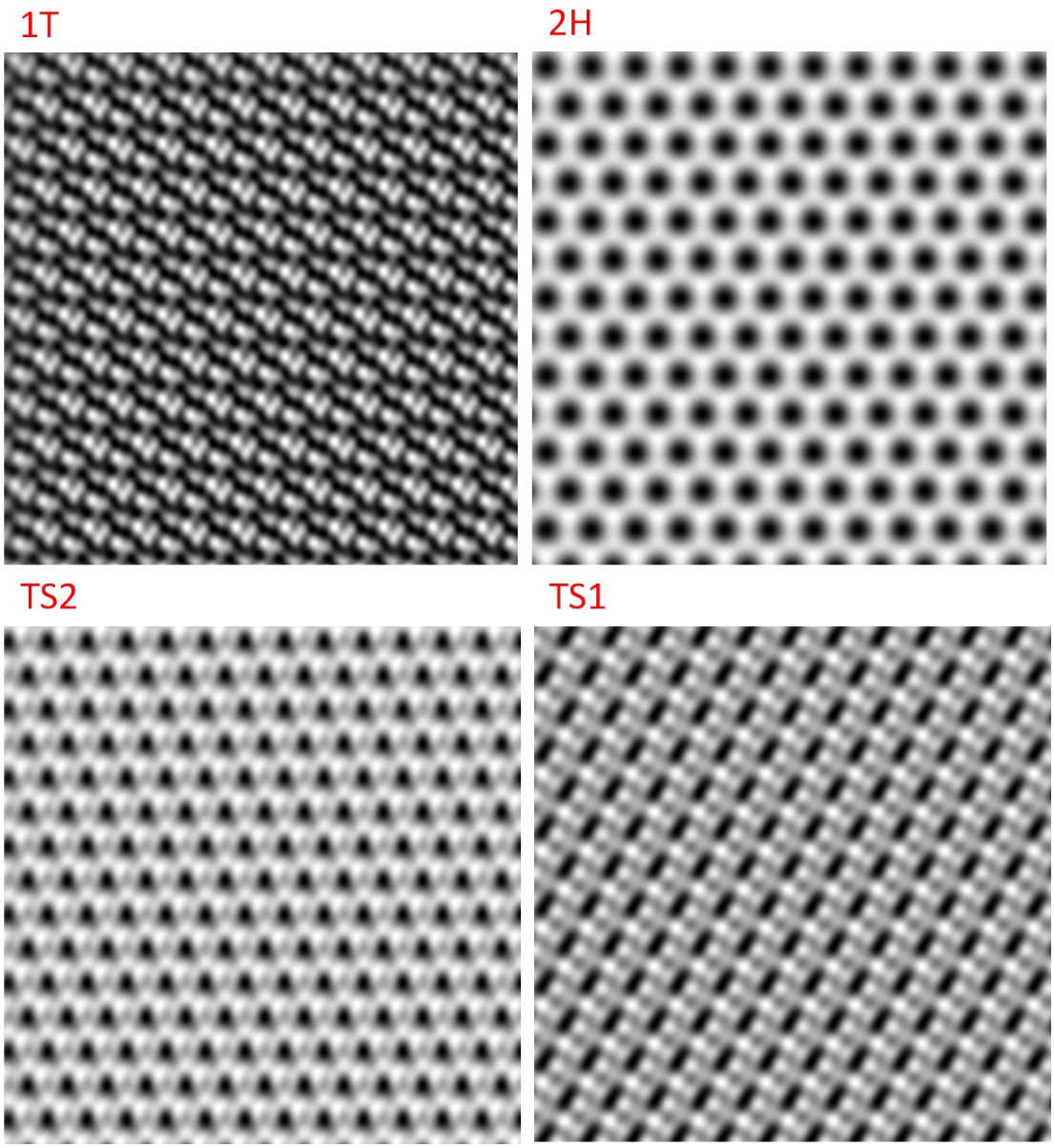


Figure S9. TEM image simulation based on atomic model of single layer MoS_2 (Ovito Software) of different phases.

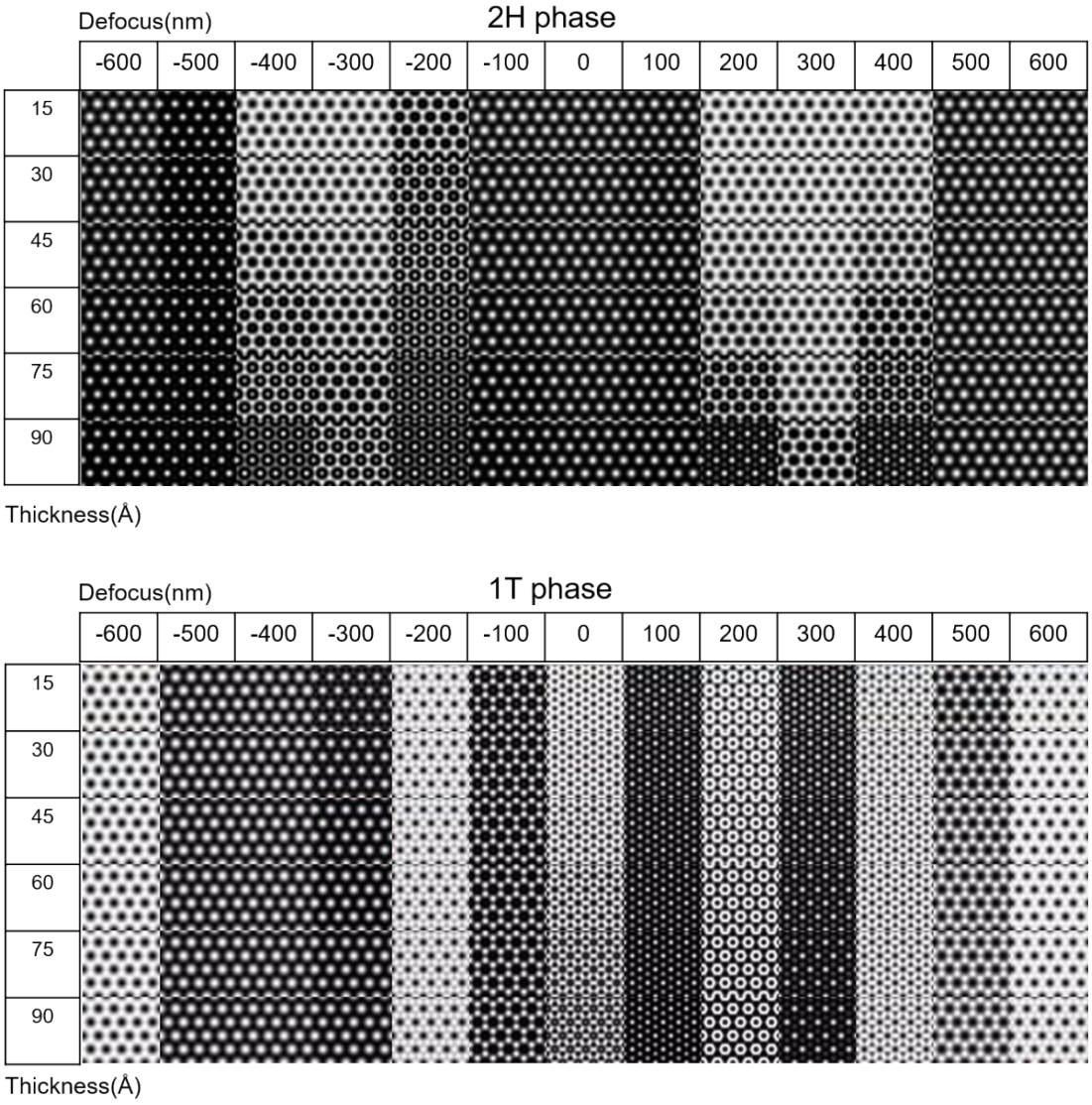


Figure S10. Focus and sample-thickness series of simulated TEM images
(Cs = 1.200 mm, convergence angle = 0.5 mrad, defocus spread = 50 nm, defocus = −5 nm.) TEM image simulation of 2H and 1T phase via MaxTempaX software, defocus range from -600nm to +600nm, layer thickness from 15nm to 90nm.

From the focus-series simulation, we can clearly see that 2H phase are matched with the experimental evidence and so does the simulation in Figure S9. On the other hand, 1T-phase focus series simulation and previous simulation (Figure S9) mismatched a little bit with the experimental images. It is because that atomic layer are ultra-thin, sulfur atoms lost its contrast at low current density. Also, the astigmatism may also cause image difference.

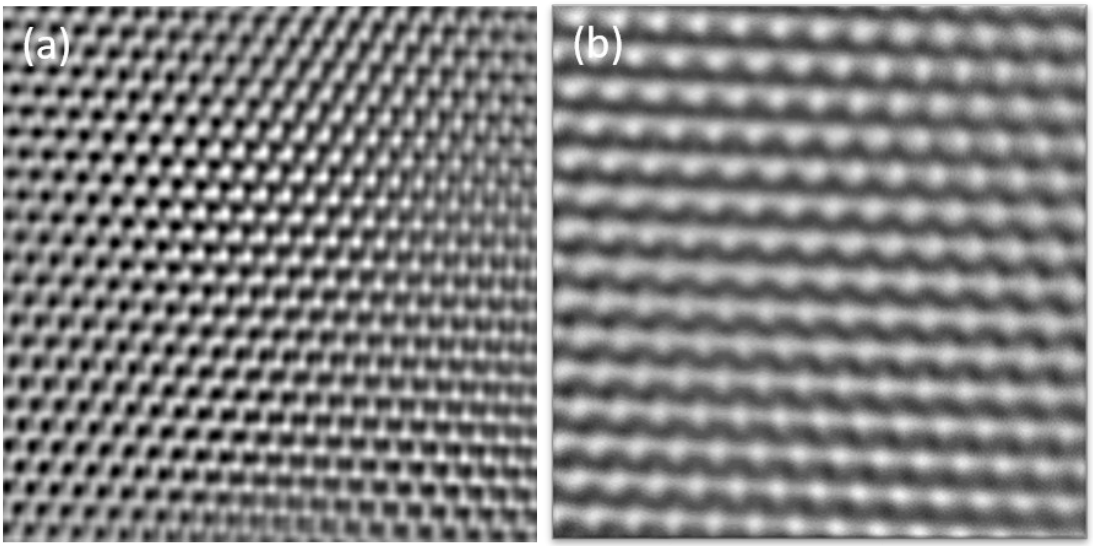


Figure S11 Two types of contrast induced from ripples.

(a) Blurred contrast while maintaining hexagonal pattern.

(b) Only one-directional contrast can be seen due to deviation of height in ripples.

Table S1. Ground state energy differences between monolayer phases of MoS_2[1]

Phase	Energy
2H	U=0 ground state
1T’	U=0.55 eV
1T	U=0.85 eV
TS1	U=1.88 eV
TS2	U=2.25 eV

1. Ryzhikov, M. R.; Slepkov, V. A.; Kozlova, S. G.; Gabuda, S. P.; Fedorov, V. E., Solid-State Reaction as a Mechanism of 1T ↔ 2H Transformation in MoS2 Monolayers. *Journal of Computational Chemistry* **2015,** *36*, 2131-2134.

Movie S1(2H→1T)

The observation of the phase transformation of 2H→1T via HRTEM.

The lattice change indicated the phase transformation.

Movie S2(1T→2H)

The observation of the phase transformation of 1T→2H via HRTEM.

Different from the S.movie 1, the distorted region involved in 1T→2H transformation.

Movie S3(Model 1)

The animation of the phase transformation of 1T→TS1→2H.

Movie S4(Model 2)

The animation of the phase transformation of 1T→TS2→2H